Engineering Journal issn: 0125-8281

Article

Neural Network-based Hybrid Estimator for Estimating Concentration in Ethylene Polymerization Process: An Applicable Approach

Wachira Daosud[1,a], Mohd Azlan Hussain[2], and Paisan Kittisupakorn[3,b,*]

1 Faculty of Engineering, Burapha University, Chonburi 20131, Thailand
2 Department of Chemical Engineering, University of Malaya, Kuala Lumpur 50603, Malaysia
3 Department of Chemical Engineering, Faculty of Engineering, Chulalongkorn University, Bangkok 10330, Thailand
E-mail: [a]wachira@buu.ac.th, [b]paisan.k@chula.ac.th (Corresponding author)

Abstract. Estimation of a monomer concentration of an ethylene polymerization process has been a challenging problem due to its highly nonlinear behavior and interaction among state variables. Applying of an extended Kalman filter (EKF) to provide the estimates of the concentration based on measured bed temperatures has usually been prone to errors. Here, alternatively, neural network-based hybrid estimators have been developed and classified into three structures which integrating of either EKF or Kalman filter (KF) to neural network (NN) to provide the estimates. The NNs are integrated to provide the estimates' error or concentration's estimates corresponding to individual structure for reducing the estimation error. Simulation results have shown that the hybrid estimators can provide good estimates under nominal condition and disturbance cases. However, in dealing with noises, the NN-KF hybrid estimator gives superior robustness with smooth and accurate estimated values.

Keywords: Hybrid estimator, neural network, extended Kalman filter, Kalman filter, linearization error, polymerization process.

ENGINEERING JOURNAL Volume 24 Issue 2
Received 1 October 2018
Accepted 8 January 2020
Published 31 March 2020
Online at https://engj.org/
DOI:10.4186/ej.2020.24.2.29

DOI:10.4186/ej.2020.24.2.29

1. Introduction

An ethylene polymerization process involves many challenging problems including nonlinear dynamic behavior, multivariable interactions between each state variable and unmeasurable state variables. In order to achieve desired objectives such as maximum profit or minimum cost, an effective model based control algorithm based on accurate estimates of states and parameters has to be applied. However, it has been known that some measurements i.e. concentrations are available at low sampling rate with significant time delays. To overcome this difficulty, robust state estimation is needed.

In recent years, one of most commonly used estimators for nonlinear chemical processes has been an extended Kalman filter (EKF) [1-3] which extends the Kalman filter (KF) approach to nonlinear systems by local linearization. This estimator offers good estimation performance as quoted in several literatures. Speicher et al. used an EKF to estimate the plate temperature in heavy plate rolling [4]. The EKF was validated by means of experimental data recorded during the measurement campaign. Dewasme et al. developed an EKF for various hardware sensor configurations taking account of their reliability and cost and including a possible kinetic parameter estimation [5]. The filters were validated using experimental data collected on a lab-scale bioreactor. McAuley and MacGregor used EKF as a soft sensor to estimate the melt index and polymer density values of a polyethylene reactor [6]. Due to the ease of implementation, the EKF has been used in many systems [7-9]. Nevertheless, the EKF has been shown to perform poorly for highly nonlinear systems because it is based on local linearization of dynamic models giving significant linearization error [10, 11].

To improve the accuracy of estimates for these nonlinear estimations, an alternative way is to combine the EKF with a well-known tool such as neural network (NN) in a hybrid estimation approach for reducing the error of estimation, as mentioned by Jarinah and coworkers [12]. NNs have several advantages which include distributed information processing and the inherent potential for parallel computation. The potential for processing and approximation relates to the operating data without prior knowledge of the process. NNs can be employed to be models, estimators and controllers depending on their learning data. Daosud et al. applied the NN as an inverse model which was formulated into an inverse model controller for a steel pickling process [13]. Kittisupakorn et al. demonstrated a multi-layer feedforward neural network based model predictive control for a steel pickling process [14]. Others applications of NNs for chemical process have also been investigated [15-18]. However, the main problem in NN modelling is the poor generalization because of over-fitting and under fitting of the data especially in dealing with noise which normally occur in real process [19]. To overcome the problems of the conventional NN observer, NN-based observer coupled with conventional (model-based) types are implemented [12]. Hussain et al. used a hybrid neural network (HNN) to predict porosity in a food drying process [20]. Other applications of NN-based observers can also be found in many papers [21-23]. However, for conditions where this observer is not accurate enough for the process systems, Ideal hybrid observers which are combinations of more than one observer such as NN-based observer coupled with EKF should be implemented for improving estimation in certain systems [12]. Nevertheless, NN-based hybrid estimators have rarely been reported with any applications so far [24].

In this paper, a novel NN-based hybrid estimator has been studied and investigated for application to an ethylene polymerization process considered as one of biggest synthetic commodity sources of polymer in the world today. The highly nonlinear dynamic behavior, its multivariable nature and the interaction between state variables of the process lead to the difficulty in estimating unmeasurable states by both KF and EKF. Therefore, the aim and contribution of this work are to develop NN-based hybrid estimators combining of NN and KF or EKF for estimating monomer concentration using available bed temperatures. To demonstrate the robustness of the proposed estimators, tests involving various disturbances and noise have been performed in these studies.

2. Process Description

An ethylene polymerization reactor is a fluidized bed reactor which uses feed gas partly combined with the recycled gas to introduce bubbling fluidized bed of Ziegler-Natta catalysts. The reactor consists of a catalyst feeder, a product withdrawal system which is employed to maintain a constant bed height inside the reactor, gas recycling which includes a cyclone and a compressor as shown in Fig. 1. Gaseous species are Ethylene (monomer), Butene (comonomer), hydrogen and nitrogen. Mathematical models of the ethylene polymerization reactor studied in [25] is used in this work. Physical properties and nominal operating of the system are summarized in Table 1.

Mass and energy balance equations of the process (Eq. (1)-(7)) indicate the behavior of state variables changing with time are as follows.

Mass balances on gaseous species:

$$V_g \frac{dC_{M1}}{dt} = F_{M1} - x_{M1}B_t - C_{M1}Y_c k_{p1} e^{-\frac{E}{R}(1/T-1/T_{ref})} \quad (1)$$

$$V_g \frac{dC_{M2}}{dt} = F_{M2} - x_{M2}B_t - C_{M2}Y_c k_{p2} e^{-\frac{E}{R}(1/T-1/T_{ref})} \quad (2)$$

$$V_g \frac{dC_H}{dt} = F_H - x_H B_t \quad (3)$$

$$V_g \frac{dC_N}{dt} = F_N - x_N B_t \quad (4)$$

Catalyst active sites balance:

$$\frac{dY_c}{dt} = F_c a_c - k_d Y_c - O_p Y_c / B_w \tag{5}$$

Reactor energy balance:

$$\begin{aligned}&(M_r Cp_r + B_w Cp_p)\frac{dT}{dt}\\&= (F_{M1}Cp_{M1} + F_{M2}Cp_{M2} + F_H Cp_H + F_N Cp_N)(T_f - T_{ref})\\&+ F_g Cp_g (T_g - T_{ref}) - M_{w1} C_{M1} Y_c \Delta H_r k_{p1} e^{-\frac{E}{R}(1/T - 1/T_{ref})}\\&-(F_g + B_t)Cp_g (T - T_{ref}) - O_p Cp_p (T - T_{ref})\end{aligned} \tag{6}$$

Heat exchanger energy balance:

$$M_g Cp_g \frac{dT_g}{dt} = F_g Cp_g (T_{gi} - T_g) + F_w Cp_w (T_{wi} - T_{wo}) \tag{7}$$

The above equations are linearized and used in the EKF algorithm consisting of seven state variables which are C_{M1}, C_{M2}, C_H, C_N, Y_c, T and T_g. In addition, they are used to provide simulated process data which will be divided into three sets: training, testing and validating data. Use of each model equations to design different estimation approaches will be described in the next section.

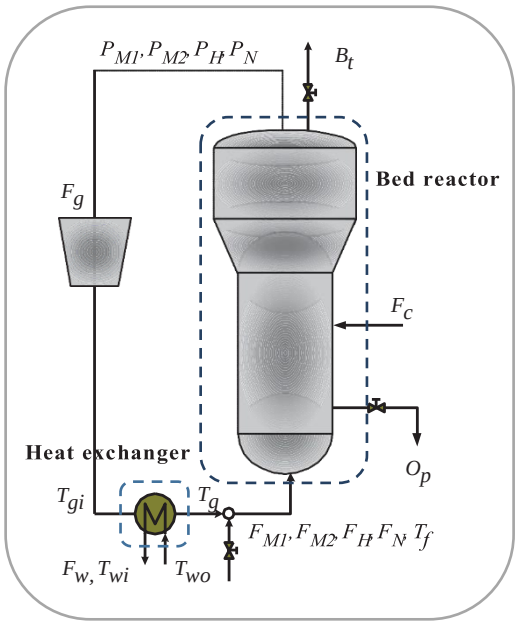


Fig. 1. The ethylene polymerization reactor.

3. Design of Estimators

In general, chemical reactions can be observed from measurements of the amount of reactants used which are the concentrations of monomers in this process. However, they cannot be directly measured, so an estimation approach is needed. An extended Kalman Filter (EKF) consists of two computational steps, i.e., corrector and predictor steps. Thereby equations used in the EKF are divided into two major parts which are time update equations and measurement update equations used in the predictor and corrector steps respectively. One important factors affecting the estimated accuracy is the model's accuracy; If it represents a process well, the estimate certainly is accurate. In this work, the linearized models used in the prediction step of the EKF is the discrete-time state model based on mathematical process models (Eq. (1)-(7)) in the form:

$$x_{k+1|k} = f(x_{k|k}, u_k) \tag{8}$$

where $x_{k|k}$ is the state vector at time k and u_k is the input vector at time k ([$F_{m1}(k)$, $F_{m2}(k)$, $F_H(k)$, $F_N(k)$, $F_c(k)$])

The priori estimate error covariance ($P_{k+1|k}$) is calculated from Eq. (9)

$$P_{k+1|k} = A_k \cdot P_{k|k} \cdot A_k^T + Q \tag{9}$$

where A_k is the nxn dimension state transition matrix at time k, obtained from solving the differential process model equations according to each state variable ($A_k = \frac{\partial f(x,u)}{\partial x}|_{x_{k|k}, u_k}$) and Q is the nxn dimension process error covariance matrix. The estimated measurement equation is defined as

$$y_{k+1|k} = c(x_{k+1|k}) \tag{10}$$

where $y_{k+1|k}$ is the estimated measurement variable as $T_{k+1|k}$.

In the correction step, the equations consist of a gain of Kalman, posteriori estimate error covariance and a state estimated equation, Eq. (11) – (13), respectively.

$$K_{est,k+1} = P_{k+1|k} C_{k+1}^T \left(C_{k+1} P_{k+1|k} C_{k+1}^T + R\right)^{-1} \tag{11}$$

$$P_{k+1|k+1} = \left(I - K_{est,k+1} C_{k+1}\right) P_{k+1|k} \tag{12}$$

$$x_{k+1|k+1} = x_{k+1|k} + K_{est,k+1}\left(\varepsilon_{k+1}\right);\ \varepsilon_{k+1} = y_{k+1} - y_{k+1|k} \tag{13}$$

where C_{k+1} is the 1xn dimension measurement matrix defined as $C_{k+1} = \frac{\partial c(x,u)}{\partial x}|_{x_{k+1|k}}$, y_{k+1} is measurement value at time k+1 and R is the measurement noise covariance.

To estimate the monomer concentration (C_{m1}), the EKF uses information of measured bed temperature (T_{mea}) at each sampling time step to determine the states and parameters based on the Eq. (8)-(13). Since the ethylene polymerization process is complicated and highly non-linear, the EKF usually provides less accurate estimates of states. The prediction step equations have been formulated as follows:

DOI:10.4186/ej.2020.24.2.29

$$x_{k+1|k} = A_k \cdot x_{k|k} + B_k \cdot u_k \tag{14}$$

where $A_k = \frac{\partial f(x,u)}{\partial x}|_{x_{k|k},u_k} = \begin{bmatrix} \frac{\partial f_1}{\partial x_1} & \cdots & \cdots & \frac{\partial f_1}{\partial x_7} \\ \frac{\partial f_2}{\partial x_1} & \cdots & \cdots & \frac{\partial f_2}{\partial x_7} \\ \vdots & \ddots & \ddots & \vdots \\ \frac{\partial f_7}{\partial x_1} & \cdots & \cdots & \frac{\partial f_7}{\partial x_7} \end{bmatrix}_{x_{k|k},u_k}$

and $B_k = \frac{\partial f(x,u)}{\partial u}|_{x_{k|k},u_k} = \begin{bmatrix} \frac{\partial f_1}{\partial u_1} & \cdots & \frac{\partial f_1}{\partial u_5} \\ \frac{\partial f_2}{\partial u_1} & \cdots & \frac{\partial f_2}{\partial u_5} \\ \vdots & \ddots & \vdots \\ \frac{\partial f_7}{\partial u_1} & \cdots & \frac{\partial f_7}{\partial u_5} \end{bmatrix}_{x_{k|k},u_k}$

Simulation results (Fig. 4(a)) have shown that the estimates by the EKF based on linearized models which are normally obtained from basic identification method, cannot fit to the target data (monomer concentration). In the same way, using a single NN estimator (NNest) for highly non-linear process provided the estimated result with less accuracy especially in dealing with noise in various conditions as shown in Table 3 for mean squared error (MSE). The components of NNest are defined in Table 2. For improving of the estimated accuracy, the estimation approach is introduced in hybrid form which is NN cooperating with EKF.

In this work, three hybrid estimators: NNEKF1, NNEKF2 and NNKF corresponding to model format and inside data flow are proposed to provide estimates of the monomer concentration (C_{m1}) in the reactor of the ethylene polymerization process, the estimators are applied to provide C_{m1} from the measured bed temperature (T_{mea}) at each sampling time. The details of each estimator are described as follows.

Table 1. The physical properties and nominal operating conditions of the ethylene polymerization reactor [25].

Concentration (mol/m^3)		Deviation rate constant (1/s); k_d	0.0
Monomer; C_{M1}	297.06	Temperature (K)	
Co-monomer; C_{M2}	116.17	Bed; T	355.85
Nitrogen; C_N	166.23	Feed; T_f	300.15
Hydrogen; C_H	105.78	Reference; T_{ref}	360.15
Heat capacity (J/mol·K)		Recycle stream before cooling; T_{gi}	409.15
Monomer; Cp_{M1}	46.05	Recycle stream after cooling; T_g	324.85
Co-monomer; Cp_{M2}	100.48	Cooling water before cooling; T_{wi}	293.15
Nitrogen; Cp_N	28.89	Cooling water after cooling; T_{wo}	308.15
Hydrogen; Cp_H	32.24	Activation energy for propagation (J/mol); E	37681.2
Recycle gas and water; Cp_w	75.36	Water holdup in the heat exchanger (mol); M_w	2.0x10^6
Heat capacity of polymer (J/g·K); Cp_p	3.56	Thermal capacitance of the vessel (kJ/K); M_rCp_r	5.86x10^7
Flow rate (mol/s)		Polymer outlet rate (kg/s); O_p	3.64
Cooling water; F_w	3.11x10^5	Ideal gas constant (atm·m^3/K·mol); R	82.6x10^{-6}
Recycle; F_g	8500	Number of moles of catalyst site (mol); Y_c	5.85
Monomer; F_{M1}	131.13	Heat of reaction (J/g); ΔH_r	-3743
Co-monomer; F_{M2}	3.51	Volume of gas phase in the reactor (m^3); V_g	500
Nitrogen; F_N	2.52	Rate constant (m^3/mol·s)	
Hydrogen; F_H	1.60	Monomer Propagation; k_{p1}	0.085
Bleed; B_f	10.39	Co-monomer Propagation; k_{p2}	0.003
Catalyst flow rate (kg/h); F_c	2.0		
Pressure (atm)			
Total pressure; P_t	20.0		
Active site concentration (mol/kg); a_c	0.55		
Mass of polymer in the bed (kg); B_w	7.0x10^5		

3.1. NNEKF1 Estimator

The first estimator, NNEKF1, is developed from EKF combined with NN, which is an idea of reducing the error between EKF estimated value and the plant value, as shown in Fig. 2. Due to the non-linearity of the process to be estimated and the observability of measured variables, an EKF is applied to provide the first estimate of monomer concentration (C_{est1}) in the NNEKF1 estimators. After that the C_{est1} is included along with the different value (Er) between the monomer concentration from the plant (C_{m1}) and the first estimate of the monomer concentration from the EKF (C_{est1}) where Er is predicted by the NN1. Then the output of the NNEKF1 estimator

is the second estimated monomer concentration (C_{est2}) obtained from the summation of C_{est1} and Er.

For EKF in this structure, the equations used in the prediction step are state space form equations, linearized model, as defined in Eq. (14). The A and B matrixes are updated corresponding to the estimated state variables ($x_{k+1|k+1}$) in the correction step Eq. (11)-(13). The priori estimate error covariance ($P_{k+1|k}$) is obtained from Eq. (9). The C_{est1} is determined by the EKF according to the measured bed temperature input (T_{mea}). After that C_{est1} and T_{mea} are used by the NN1 for predicting the error (Er). The NN1 is designed as a feedforward neural network with the optimal structure consisting of 3 nodes in input layer, 4 nodes in the one hidden layer and one output node with respect to the minimum mean squared error (MSE) method [13]. The optimal structure of NN (NN1) is shown in Fig. 2.

The selected inputs to the NN are the previous and current values of C_{est1} and the current value of T_{mea}, which directly correspond to the current value of Er or the output of the network. The training, testing and validation data sets are obtained by selecting appropriate data from the output of EKF (C_{est1}), the measured bed temperature (T_{mea}) and the monomer concentration (C_{m1}) obtained from the simulation of the ethylene polymerization process models by solving Eq. (1)–(7). These equations are solved to obtain the process states according to various step changes in the input variables, i.e., flow rates (F_{m1}, F_{m2}, F_H, F_N and F_c). The target data for NN1 training or the errors (Er) are produced from the different value of C_{m1} and C_{est1}. The defined NN is trained with the Levenberg–Marquardt algorithm in the MATLAB NN toolbox where the objective is to reduce the error between the NN predicted error and the actual error value. The optimal structure is selected by the minimum MSE method which provides the NN structure giving the minimum MSE value in prediction. Details of the procedure for obtaining feedforward neural network models are explained in [13]. The optimal NN (3-4-1 configuration), NN1, is connected in series after the EKF to predict the Er at current time. Then the Er is added to C_{est1} for improving the estimated accuracy and sent out as NNEKF1 estimator output, the second estimated monomer concentration (C_{est2}). The component of the NN1 is shown in Table 2.

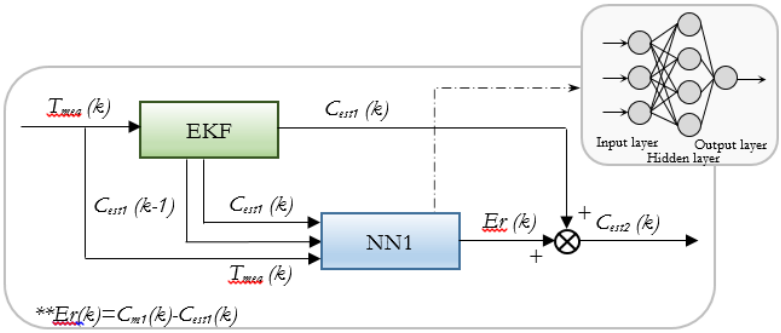


Fig. 2. The structure of NNEKF1 estimator.

Table 2. The components of NN models.

Components	NNest	NN1	NN2	NN3
Training Algorithm	Levenberg–Marquardt algorithm			
Basis Function	Linear Basis Function			
Activation Function	Sigmoid Function			
Input of NN	$T_{mea}(k)$, $T_{mea}(k\text{-}1)$, $T_g(k)$, $T_g(k\text{-}1)$	$T_{mea}(k)$, $C_{est1}(k)$, $C_{est1}(k\text{-}1)$	$x_{i,k+1\|k+1}$; $i=1,2,...,7$	$\hat{x}_{i,k+1\|k+1}$; $i=1,2,...,7$
Output of NN	$C_{est}(k)$	$Er(k)$	$C_{est}(k)$	$C_{est}(k)$
Optimal structure (Input node-Hidden node-Output node)	4-4-1	3-4-1	7-4-1	7-10-1

3.2. NNEKF2 Estimator

The structure of this estimator is designed by combining between EKF and a feedforward neural network as shown in Fig. 3(a) (NN2). The models of EKF are the same as in the NNEKF1 estimator. The EKF is used to estimate all state variables of the process based on the models at current time. From the Fig. 3(a), it is realized that as the EKF estimates both monomer concentration and bed temperature based on measured bed temperatures, the estimated bed temperature (T_{est}) is more accurate than the estimated concentration (C_{est1}). From this advantage,

the T_{est} or $T_{k+1|k+1}$ and the other estimated state variables are continually used to find accurate predicted monomer concentration (C_{est}) by NN.

The inputs of NN2 are selected as the current value of all estimated state variables from the EKF in relative with the current value of T_{mea}. The training target data are simulated monomer concentration (C_{m1}), by solving Eqs. (1)–(7) at the current time. The training and obtaining procedure of the optimal NN are defined in [12]. The optimal NN of the NNEKF2 estimator, NN2, consists of 7 nodes of input layer, 4 nodes of one hidden layer and one node of output layer (7-4-1 configuration). The optimal NN is used to predict the monomer concentration which is the output of the NNEKF2 estimator from the estimated state variables of EKF. Details regarding NN2 are defined in Table 2.

3.3. NNKF Estimator

From the previous section, it is observed that each estimated state variable obtaining from the EKF is the state variable of the process. Therefore, all estimated state variables are directly affected by the error of measurement and noise signal as defined in Eq. (13). Also, they are very sensitive because of the changing of A and B matrix in every time state as shown in Eq. (14).

For this reason, the estimates of C_{est1} are low accuracy. When they are sent as the inputs to NN, the NN will give low accuracy as well. To handle this, another estimator is designed using a KF based on state space model (linearized model) of the process combined with NN as shown in Fig. 3(b). The KF uses the time update equations in the form of state space model obtaining from subspace method via MATLAB identification toolbox. The state space model is indicated as follows

$$\hat{x}_{k+1|k} = A \cdot \hat{x}_{k|k} + B \cdot u_k \quad (15)$$

$$y_{k+1|k} = C \cdot \hat{x}_{k+1|k} \quad (16)$$

where $\hat{x}_{k|k}$ is *pseudo* state vector at time k and $y_{k+1|k}$ is the estimated measurement variable as $T_{k+1|k}$.

Simulation of the ethylene polymerization process under the condition as Table 1 is carried out to provide data for training and testing of the proposed estimators The *pseudo* state variables consist of seven state values that are selected by maximum percentage of fitting via subspace method and also based on process models. The sensitivity analysis, in case of noise, indicates that the estimated *pseudo* state variables are less sensitive than the estimated state variables. Although random noise is introduced more than 5 % of the measured value (300 K), the third *pseudo* state variable is almost identical to its normal condition.

With less effect of noise and measurement error on the estimated *pseudo* state variables, the NN can provide the accurate estimated monomer concentration according to T_{mea}. The NN in the NNKF estimator (NN3) consists of 7 nodes of input layer, 10 nodes of one hidden layer and one node of output layer (7-10-1 configuration). The inputs of NN3 are selected as the current value of all estimated *pseudo* state variables from the KF in relative with the current value of T_{mea}. Hence, the estimated monomer concentration is predicted by NN3, which is the output of NNKF estimator. The components of NN3 are defined in Table 2.

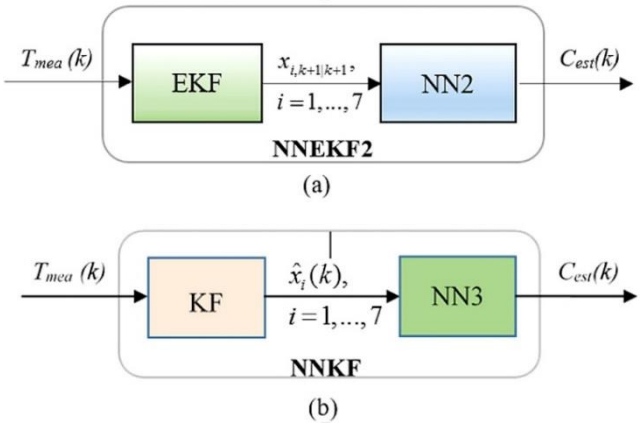


Fig. 3. The structure of estimators: a) NNEKF2 and b) NNKF.

4. Simulation Results

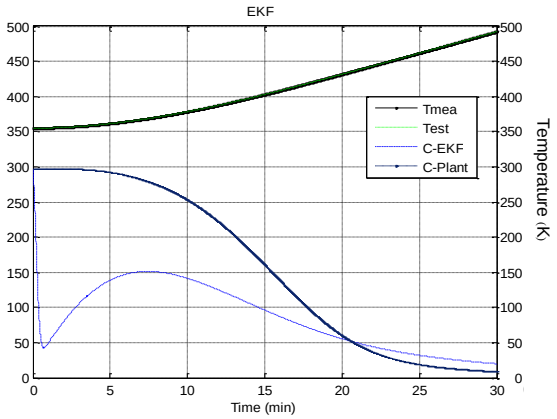
All proposed estimators are applied and compared to estimate the monomer concentration in the ethylene polymerization reactor using the measured bed temperature at each sampling time. The performances and robustness tests are carried out with four cases: the nominal case, noise case, disturbance case and disturbance together with noise case.

4.1. Nominal Case

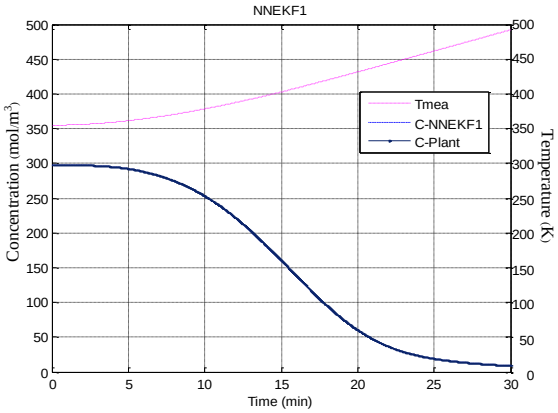
In this case, the estimators provide the estimated monomer concentration using the measured bed temperatures which are calculated from the ethylene polymerization process models under the nominal condition. The simulation results of all estimators are shown in Fig. 4.

From Fig. 4(a), with the measured bed temperature, the EKF gives poor estimates of the monomer concentration as indicated in the dash line. Because of the complexity of the process, highly non-linear behavior and interaction between state variables (i.e. C_{m1} and T), the state space equations in the predictor step of EKF rarely represent the exact states of the process. So, the predictor step provides the predicted state variables with high error which strongly effect to the estimated monomer concentration in the corrector step where the function of Kalman gain and measurement error are combined (eq. (13)). However, the NNEKF1 provides good estimates because of the error compensation by the combined NN as shown in Fig. 4(b). For the NNEKF2 and NNKF

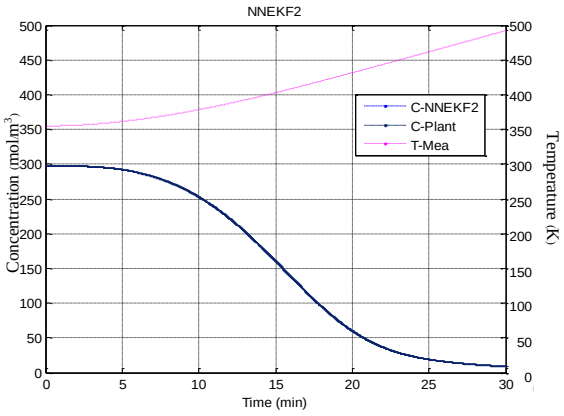
estimators (Fig. 4(c) and (d)), the estimated monomer concentration are provided in a satisfied accuracy with less MSE as shown in Table 3 because accurate estimated bed temperature ($T_{k+1|k+1}$) is used in NN for estimating the concentration.



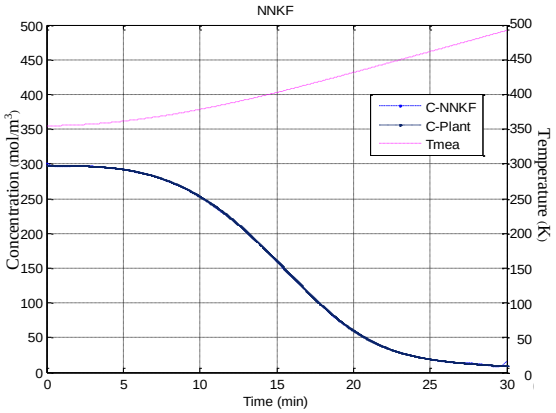
(a)



(b)



(c)



(d)

Fig. 4. The estimated monomer concentration under normal condition: (a) EKF, (b) NNEKF1, (c) NNEKF2 and (d) NNKF.

4.2. Noise Case

For the case in the presence of noise, random noises accounting to 2 K from the bed temperature are introduced into the measured bed temperature to evaluate robustness and performance of the estimators under real situations. The estimated results show that the NNEKF1 and NNEKF2 cannot provide the estimates of the monomer concentration due to the inputs to NN (i.e. the first estimated monomer concentration) are strongly affected by noise signal which directly cause inaccurate estimated concentration in the further NN estimation. On the other hand, the NNKF estimator excellently estimates the monomer concentration under presence of noise of 2 K till even 35 K as shown in Fig. 5(a) and 5(b), respectively. Since estimated *pseudo* state variables continually fed into NN are slightly affected by noise signal, estimated concentration by NN are reasonably accurate. The MSE values of estimations using these estimators in dealing with noises are summarized in Table 3.

Table 3. Performance comparison between the designed estimators under the nominal case, noise case and disturbance case.

Condition	MSE values			
	NNest	NNEKF1	NNEKF2	NNKF
Nominal case	4.732	1.124×10^{-3}	2.922×10^{-5}	1.632×10^{-4}
Noise case (2K)	10.849	NaN†	NaN	1.633×10^{-4}
Noise case (35K)	2599.951	NaN	NaN	2.532×10^{-4}
Disturbance case (F_{m1})	56.833	48.091	53.067	50.861
Disturbance case (F_c)	4.864	9.870×10^{-2}	216.555	1.174×10^{-1}
Disturbance (F_{m1} and F_c) with noise (35K)	2462.166	NaN	NaN	47.152

†NaN = the estimator cannot estimate the concentration.

4.3. Disturbance Case

The designed estimators are also tested involving several disturbances. For the disturbance case, changes in the flow rate of the streams, F_{m1} and F_c are introduced by increasing its nominal operation values by 10%. Fig. 6 shows the results of the NNEKF1, NNEKF2 and NNKF estimators for estimating the monomer concentration with introduced F_{m1} disturbance. It can be seen from Fig. 6 that all of the proposed estimators can give the estimates of the monomer concentration accurately. Their results show the small discrepancy from the target concentration along the time as shown in Fig 6(a), 6(b) and 6(c). Similarly, the estimated results in the presence of changes in F_c have less error indicating from their MSE values. The MSE values obtained from the NNEKF1, NNEKF2 and NNKF estimators for this case are summarized in Table 3. They indicate that these estimators can still provide good performances even when the disturbances are introduced into the system.

4.4. Disturbance with Noise Case

In this case, F_{m1} and F_c are introduced by increasing their nominal operation values by 10% together with random values of noise by 35 K from the bed temperature to the process. From the estimated results show that, there is only one hybrid estimator which is NNKF can handle this condition. Due to the fact that the introduced measurement noise can be handled by KF leading to accurate estimated *pseudo* state variable, therefore the NN can provide good estimates of the concentration. It can be seen that NNKF not only provides the good estimated accuracy of the monomer concentration as shown in Fig. 7, it gives the smooth estimated result and less MSE value as well (Table 3).

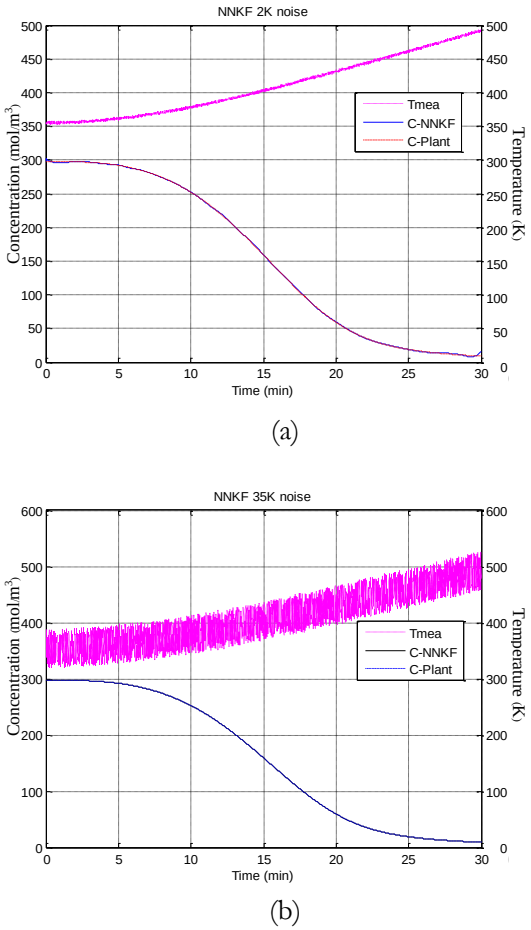
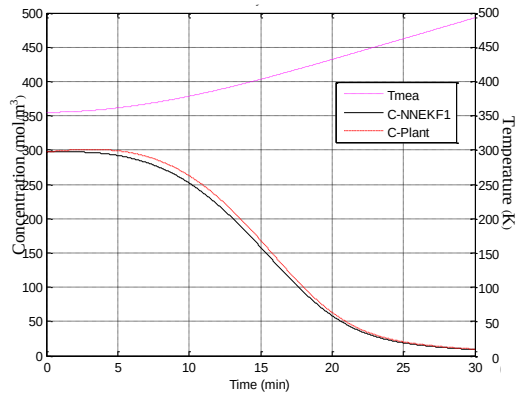
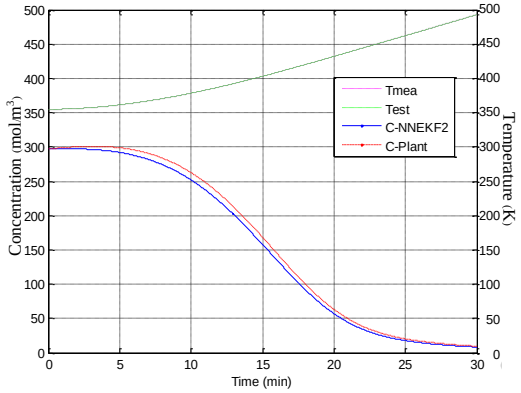


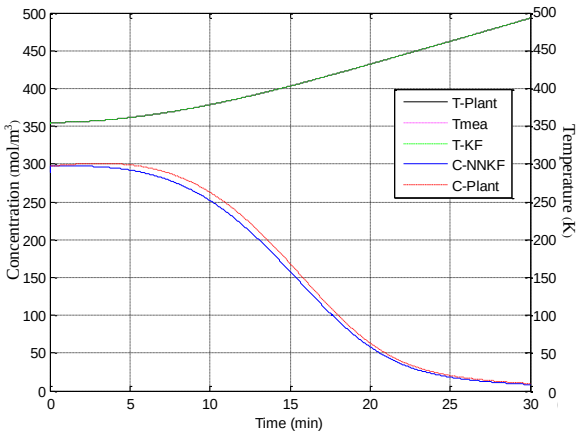
Fig. 5. The estimated monomer concentration under presence of noise: (a) 2K noise NNKF and (b) 35K noise NNKF.



(a)



(b)



(c)

Fig. 6. The estimated monomer concentration under the disturbance case: (a) NNEKF1, (b) NNEKF2; (c) NNKF.

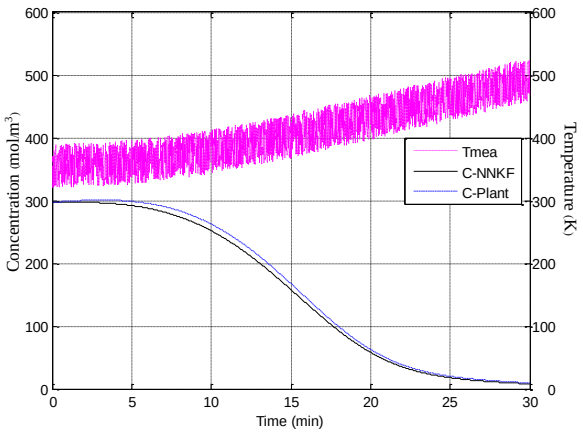


Fig. 7. The estimated monomer concentration under the disturbance and noise case by using NNKF estimator.

5. Conclusions

Most chemical processes are complex, highly nonlinear with multivariable interacting systems, such as the ethylene polymerization process. Therefore, they are normally difficult to observe current conditions or state variables based on available measurements. Hence, in this work, a novel method using hybrid neural network (HNN) with Kalman filter (KF) is designed with three structures according to inside models and data flows for estimating the monomer concentration of an ethylene polymerization reactor using the measured bed temperature. Simulation study has shown that all proposed hybrid estimators give good estimates of monomer concentration under nominal and with disturbances conditions with low MSE values. However, the combined KF and NN estimator using the linearized model and pseudo state as NN input, provides superior estimation in dealing with noises as where providing the accurate and smooth estimated monomer concentration. These results validate the robustness of the NNKF hybrid estimator which make it highly promising to be implemented in nonlinear multivariable interacting systems such as this ethylene polymerization process.

Acknowledgement

The financial support to this work under the Research Grant of Burapha University through National Research Council of Thailand (Grant no. 220/2561), Rachadapisek Sompote Fund of Chulalongkorn University for Research Unit on Intelligent Control Automation of Process Systems and Chulalongkorn Academic Advancement into Its 2nd Century Project (Smart Energy).

Nomenclature

a_c	active site concentration of catalyst (mol/kg)
B_w	mass of polymer in the fluidized bed (g)
B_t	overhead gas bleed (mol/s)

C_{M1} concentration of monomer (mol/m^3)
C_{M2} concentration of co-monomer (mol/m^3)
C_N concentration of nitrogen (mol/m^3)
C_H concentration of hydrogen (mol/m^3)
Cp_{M1} heat capacity of monomer (J/mol K)
Cp_{M2} heat capacity of co-monomer (J/mol K)
Cp_H heat capacity of hydrogen (J/mol K)
Cp_N heat capacity of nitrogen (J/mol K)
Cp_w heat capacity of recycle gas and water (J/mol K)
Cp_p heat capacity of polymer (J/mol K)
E activation energy for propagation (J/mol)
F_c catalyst flow rate (kg/s)
F_w cooling water flow rate (mol/s)
F_g recycle flow rate (mol/s)
F_{M1} monomer flow rate (mol/s)
F_{M2} co-monomer flow rate (mol/s)
F_H hydrogen flow rate (mol/s)
F_N nitrogen flow rate (mol/s)
i state variable i of the process
j component j in the reactor
k_d deactivation rate constant (1/s)
k_{p1} monomer propagation rate constant (m^3/mole s)
k_{p2} co-monomer propagation rate constant (m^3/mole s)
M_rCp_r thermal capacitance of the reaction vessel (kJ/K)
O_p polymer outlet rate (kg/s)
R ideal gas constant (atm m^3/K mol)
T bed temperature (K)
T_f feed temperature (K)
T_{ref} reference temperature (K)
T_{gi} temperature of recycle stream before cooling (K)
T_g temperature of recycle stream after cooling (K)
T_{wi} cooling water temperature before cooling (K)
T_{wo} cooling water temperature after cooling (K)
V_g volume of gas phase in the reactor (m^3)
Y_c number of moles of catalyst site (mol)
ΔH_r heat of reaction (J/g)

References

[1] A. Gopalakrishnan, N. S. Kaisare, and S. Narasimhan, "Incorporating delayed and infrequent measurements in Extended Kalman Filter based nonlinear state estimation," *J Process Contr*, vol. 21, pp. 119-129, Jan. 2011.

[2] G. R. Jonsson, S. Lalot, O. P. Palsson, and B. Desmet, "Use of extended Kalman filtering in detecting fouling in heat exchangers," *Int J Heat Mass Tran*, vol. 50, pp. 2643-2655, Jul. 2007.

[3] S. Kim, K. Y. Kim, and C. H. Huang, "Estimation of time-dependent reaction coefficient with extended Kalman filter approach," *J Ind Eng Chem*, vol. 18, pp. 349-354, Jan. 2012.

[4] K. Speicher, A. Steinboeck, A. Kugi, D. Wild, and T. Kiefer, "Analysis and design of an Extended Kalman Filter for the plate temperature in heavy plate rolling," *J Process Contr*, vol. 24, pp. 1371-1381, Sep. 2014.

[5] L. Dewasmea, G. Goffaux, A. L. Hantson, and A. V. Wouwer, "Experimental validation of an Extended Kalman Filter estimating acetate concentration in E. coli cultures," *J Process Contr*, vol. 23, pp. 148-157, Feb. 2013.

[6] K. B. McAuley and J. F. MacGregor, "On-line inference of polymer properties in an industrial polyethylene reactor," *AICHE J*, vol. 37, pp. 825-835, 1991.

[7] R. Huangc, S. C. Patwardhan, and L. T. Biegler, "Robust stability of nonlinear model predictive control based on extended Kalman filter," *J Process Contr*, vol. 22, no. 1, pp. 82-89, Jan. 2012.

[8] H. Khodadadi and H. Jazayeri-Rad, "Applying a dual extended Kalman filter for the nonlinear state and parameter estimations of a continuous stirred tank reactor," *Comput Chem Eng*, vol. 35, no. 11, pp. 2426-2436, Nov. 2011.

[9] M. Mauricio-Iglesias, K. V. Gernaey, and J. K. Huusom, "State Estimation in Fermentation of Lignocellulosic Ethanol. Focus on the Use of pH Measurements," *Comput-Aided Chem En*, vol. 37, pp. 1769-1774, 2015.

[10] F. Daum, "Nonlinear filters: beyond the Kalman filter," *IEEE Aero El Sys Mag*, vol. 20, pp. 57-69, 2005.

[11] J. Prakash, S. Patwardhan, and S. Shah, "Constrained nonlinear state estimation using ensemble Kalman filter," *Ind Eng Chem Res*, vol. 49, pp. 2242-2253, 2010.

[12] J. M. Ali, N. H. Hoang, M. A. Hussain, and D. Dochain, "Review and classification of recent observers applied in chemical process systems," *Comput Chem Eng*, vol. 76, pp. 27-41, May 2015.

[13] W. Daosud, P. Thitiyasook, A. Arpornwichanop, P. Kittisupakorn, and M. A. Hussain, "Neural network inverse model-based controller for the control of a steel pickling process," *Comput Chem Eng*, vol. 29, no. 11, pp. 2110-2119, Sep. 2005.

[14] P. Kittisupakorn, P. Thitiyasook, M. A. Hussain, and W. Daosud, "Neural network based model predictive control for a steel pickling process," *J Process Contr*, vol. 19, no. 4, pp. 579-590, Apr. 2009.

[15] A. Arpornwichanop and N. Shomchoam, "Control of fed-batch bioreactors by a hybrid on-line optimal control strategy and neural network estimator," *Neurocomputing*, vol. 72, no. 10-12, pp. 2297-2302, Jun. 2009.

[16] W. Daosud, K. Jariyaboon, P. Kittisupakorn, and M. A. Hussain, "Neural network based model predictive control of batch extractive distillation process for improving purity of acetone," *Engineering Journal*, vol. 20, no. 1, pp. 47-59, Jan. 2016.

[17] M. Ghasem, S. A. Sata, and M. A. Hussain, "Temperature control of a bench-scale batch polymerization reactor for polystyrene production," *Chem Eng Technol*, vol. 30, pp. 1193-1202, 2007.

[18] P. Kittisupakorn, T. Charoenniyom, W. Daosud, "Hybrid neural network controller design for a batch reactor to produce methyl methacrylate," *Engineering Journal*, vol. 18, no. 1, pp. 145-162, Oct. 2013.

[19] J. M. Ali, M. A. Hussain, M. O. Tade, and J. Zhang, "Artificial intelligence techniques applied as estimator in chemical process systems – A literature survey," *Expert Syst Appl*, vol. 42, no. 14, pp. 5915-593, Aug. 2015.
[20] M. A. Hussain, M. S. Rahman, and C. W. Ng, "Prediction of pores formation (porosity) in foods during drying: generic models by the use of hybrid neural network," *J Food Eng*, vol. 51, no. 3, pp. 239-248, Feb. 2002.
[21] C. Damour, M. Benne, L. Boillereaux, B. Grondin-Perez, and J. P. Chabriat, "NMPC of an industrial crystallization process using model-based observers," *J Ind Eng Chem*, vol. 16, no. 5, pp. 708-716, Sep. 2010.
[22] G. Porru, C. Aragonese, R. Baratti, and S. Alberto, "Monitoring of a CO oxidation reactor through a grey model-based EKF observer," *Chem Eng Sci*, vol. 55, no. 2, pp. 331-338, Jan. 2000.
[23] N. C. Wei, M. A. Hussain, A. K. A. Wahab, "Control of a batch polymerization system using hybrid neural network – First principle model," *Can J Chem Eng*, vol. 85, pp. 936-945, 2007.
[24] J. Liu, K. W. E. Cheng, and J. Zeng, "A novel multi-sensors fusion framework based on Kalman Filter and neural network for AFS application," *T I Meas Control*, vol. 37, pp. 1049-1059, 2015.
[25] S. A. Dadebo, M. L. Bell, P. J. McLellan, and K. B. McAuley, "Temperature control of industrial gas phase polyethylene reactors," *J Process Contr*, vol. 7, no. 2, pp. 83-95, 1997.

Wachira Daosud received the B.Eng. degree in chemical engineering from Burapha University (Thailand) in 1998 and the D.Eng. degree in chemical engineering from Chulalongkorn University (Thailand) in 2007.

Since 2014, she has been an Assistant Professor with the Chemical Engineering Department, Burapha University. Her research interests include modeling, dynamics and control, optimization, model predictive control, neural network and their applications in chemical and biochemical processes.

Mohd Azlan Hussain , born in Kelantan, Malaysia, received his BSc (Chemical Eng) from Sheffield University in 1980 and MSc and PhD from University of Tulsa and Imperial College in 1987 and 1996 respectively. Prof. Azlan was design engineer in Esso Production Malaysia after his BSc graduation before joining the academic field in 1985 in University of Malaya. His field of teaching and research include modelling, process controls, optimization, nonlinear control systems, instrumentation, numerical analysis, artificial intelligence techniques, control systems analysis, advanced separation system and polymerisation studies.

These activities and expertise have resulted in various outputs which include supervision/graduation of PhD, Masters student and Postdoc Researchers; 1 edited book; 9 book chapters; over 150 journal papers and 260 conference proceeding papers, as well as paper reviewers for over 80 journals. The culmination of these achievements resulted in himself being awarded the University Excellent Service Certificates and Awards in 1998, 2000, 2001, 2003, 2005, 2006, 2008, 2011, 2014, and 2016.

Paisan Kittisupakorn is a professor of Chemical Engineering at Chulalongkorn University in Bangkok, Thailand. He received his Ph.D. in Process Systems Engineering from Imperial College, University of London, United Kingdom in 1995. Since 2002, he has been awarded as an advisor for Royal Golden Jubilee Ph.D. Program with grants to support for Ph.D. students. He had also worked as an expert for National Science and Technology Development Agency, Ministry of Science and Technology, Thailand for over 8 years. Prof. Paisan currently teaches department courses in process dynamics and control, advanced process control, chemical process instrumental and control, advanced mathematics for chemical engineering. His research interests include big data application and advanced process automation: model predictive control, adaptive control neural network modeling and control, process design and synthesis, simulation and control of batch processes and batch reactors.